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THERMAL EXPANSION OF GRAPHITE

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By Peter Hidnert and W. T. Sweeney

ABSTRACT

This paper gives the results of an investigation on the thermal expansion of longitudinal and transverse sections of artificial graphite electrode (99.2 to 99.7 per cent carbon) over various temperature ranges between room temperature and 600° C.

A summary of available data by previous observers on the thermal expansion of graphite (natural and artificial) and of other forms of carbon is included.

In the present investigation the coefficients of expansion of graphite were found to be low. The transverse samples expand considerably more than the longitudinal samples (approximately 45 per cent). For both the longitudinal and transverse samples the coefficients of expansion decrease slightly as the purity (carbon content) increases. The table in the summary gives a résumé of average coefficients of expansion derived from the data on all samples for various temperature ranges between 20 and 600° C.

CONTENTS

I. INTRODUCTION

This paper gives the results of an investigation on the thermal expansion of artificial graphite, a material which finds wide applications in the industries. It is used very largely for electrodes of electric furnaces, for anodes of electrolytic cells, for the making of molds for casting metals and glass, for electrical heating elements of various sorts, for electrical contacts and arcing tips, etc. In granular or powdered form artificial graphite is used in large quantities in the manufacture of dry cells, radio batteries, and also for paint pigment, mold wash, lubricants, resistors, powder glazing, and electrotyping.

Available data by previous observers on the thermal expansion of graphite (natural and artificial) and of other forms of carbon are given in Table 1. Published values for the coefficients of expansion of various kinds of graphite vary considerably (from about 1×10^{-6} to 24×10^{-6} per ° C.).

In the present investigation expansion determinations were made over various temperature ranges between room temperature and 600° C. Five samples of artificial graphite were examined, on which a total of eight tests were made.

The authors wish to express their appreciation for the cooperation by the Acheson Graphite Co., Niagara Falls, N. Y. This company furnished the samples of graphite and information about the preparation and uses. Acknowledgment is also due to Dr. Wilmer Souder, of this bureau, for valuable suggestions.

TABLE 1.—*Summary of expansion data by previous observers on graphite and other forms of carbon*

Observer	Date	Material	Temperature or temperature range	Coefficient of linear expansion per degree centigrade	Remarks
			°C.	$\times 10^{-6}$	
Fizeau [1]	1869	Diamond	40	1.18	
		Gas carbon	40	5.40	
		Graphite, from Batougol	40	7.86	
		Anthracite, from Pennsylvania.	40	20.78	
		Coal, from Charleroy	40	27.82	
Muraoka [2]	1881	Siberian graphite (carbon content, 98.0 per cent; specific gravity, 1.8).		3.8	No relationship was found between resistance and other physical properties. Siberian graphite is the most characteristic of all the various carbons. It has the smallest electric resistance, the greatest change of resistance with temperature, and it is electronegative against all other carbons. It appears to be the best heat conductor (no quantitative measurements made). It is not distinguished by the carbon content and the specific gravity.
		Gas carbon (retorts), specific gravity, 1.80.		1.5	
		Arc carbon (specific gravity, 2.37).		1.5	
		Arc carbon of coke from the machine factory of Heilman & Co., in Mühlhausen, Germany (carbon content, 98.1 per cent; specific gravity, 1.90).		.32	
		Pencil-graphite (carbon content, 52.0 per cent; specific gravity, 2.36).		.95	
		Artificial carbon, from Kaiser and Schmidt, Berlin (carbon content, 98.0 per cent; specific gravity, 1.63).		2.05	
		Arc carbon (carbon content, 97.6 per cent; specific gravity, 1.55).		3.0	
Dewar [3]	1902	Graphite (Cumberland)			Mean coefficient of cubical expansion, 73.3×10^{-6} between $-186°$ and $+17°$ C.
Day and Sosman.[4]	1912	Artificial graphite [5] (from Acheson Graphite Co.).	0 to 200	[6] .9	The following expansion equation was derived from data on 2 samples: $10^6\beta=0.55+0.0016t$ where t is any temperature between 0° and 1,500° C. and β is the average coefficient of expansion between 0° and $t°$ C. After expansion tests the samples were usually longer than before the tests (maximum change 0.009 per cent).
			0 to 400	[6] 1.2	
			0 to 600	[6] 1.5	
			0 to 1,000	[6] 2.2	
			0 to 1,500	[6] 3.0	
Sieglerschmidt [7]	1920	Artificial graphite from A.-G. Le Carbone, Frankfurt a. M., Germany.	0 to 250	3.0	

[1] Fizeau, Pogg. Ann. d. Physik u. Chem., 18 (138), p. 26; 1869; or Compt. Rend., 68, p. 1125.
[2] Muraoka, Ann. der Physik, 13 (249), p. 307; 1881.
[3] Dewar, Proc. Roy. Soc. London, 70, p. 237; 1902; or Chem. News, 85, pp. 277, 289; 1902.
[4] Day and Sosman, J. Ind. and Eng. Chem., 4, p. 490; 1912; or J. Wash. Acad. Sci., 2, p. 284; 1912. (See also Day, Sosman, and Hostetter, Amer. J. Sci., 37 (4th series), p. 1; 1914.)
[5] C. E. Taylor, of the Acheson Graphite Co., stated (November, 1926) that this graphite was prepared from one-half inch diameter electrodes which were formed from much smaller aggregate particles than the 14-inch electrodes used in the present investigation.
[6] Computed from expansion equation given by observer.
[7] Sieglerschmidt, Mitteilungen aus dem Materialprüfungsamt (Berlin-Dahlem), 38, p. 182; 1920.

II. MATERIALS INVESTIGATED

Five samples of artificial graphite cut from the same 14-inch electrode were investigated. Samples 1180A, 1181, and 1182 were cut longitudinally and samples 1183 and 1184 were cut transversely to the direction of extrusion.

The following information relating to artificial graphite electrode material was furnished by the manufacturer:

"Green" electrode material is prepared by mixing suitable coke with a more or less plastic organic material, which upon heating carbonizes to a strong binding substance. This mixture is extruded or pressed to form "green" electrodes.

"Green" electrodes are carefully baked to carbonize as large a portion of the plastic material as possible to form strongly bound "raw" or amorphous carbon electrodes, or simply carbon electrodes.

The amorphous carbon electrodes are heated to an extremely high temperature in graphitizing furnaces, where the impurities are vaporized and distilled out and all of the carbon, both the aggregate carbon and the carbon derived from the carbonization of the plastic material, converted into artificial graphite.[1]

Table 2 gives data on the dimensions, the chemical analyses, and the densities of the samples studied. All samples were 300 mm in length, but the cross sections were varied intentionally as shown in the table.

TABLE 2.—*Chemical analyses and densities of artificial graphite*

Sample	Cross section of sample	Chemical analyses [1]			Density [4] in g/cm³ at 23° C.
		Matter soluble in water [2]	Ash	Carbon [3]	
		Per cent	*Per cent*	*Per cent*	
Longitudinal:					
1180A	22 by 10 mm	0.23	0.32	99.45	1.644
1181	Semicircle, diameter 63 mm	.19	.69	99.12	1.627
1182	do	.11	.17	99.72	1.627
Transverse:					
1183	do	.24	.60	99.16	1.635
1184 [5]	do	.18	.47	99.35	1.637

[1] Determined by T. P. Sager, of this bureau. The samples for chemical analyses were ground to pass No. 100 sieve.
[2] Matter soluble in water consisted largely of alkali chloride, sulphate, and carbonate.
[3] Per cent carbon determined by difference.
[4] Determined by E. L. Peffer, of this bureau. The volume could not be determined accurately by weighing in a liquid, because artificial graphite absorbs liquids such as water, kerosene, toluol, etc. The volume was obtained from measurements on the dimensions of the sample.
[5] For a second expansion test this sample was cut to 22 by 14 mm cross section.

III. APPARATUS

Figure 1 of Bureau of Standards Scientific Paper No. 524 (vol. 21, p. 1; 1926) shows part of the apparatus used in this investigation. With this apparatus the expansion was accurately measured by means of micrometer microscopes, which were sighted on fine wires suspended from the ends of the specimen. All expansion tests,

[1] F. Sillers, jr., of this bureau, examined a specimen of this graphite by the powder method of X ray crystal analysis. The diffraction pattern consisted of intense, definitely spaced bands, which show that the material is definitely crystalline.

except those for sample 1180A and the second test for sample 1184, were made by the method illustrated in Figure 5 of the paper mentioned. Tests for sample 1180A and the second test for sample 1184 were made by the method shown in Figure 4 of the same paper. For a detailed description of the apparatus and the methods used the reader should refer to the original paper.

IV. RESULTS ON ARTIFICIAL GRAPHITE

Since preliminary tests showed that samples of artificial graphite oxidized in air at about 640° C., the maximum temperature of the expansion tests did not exceed this temperature. The expansion data obtained on the longitudinal and transverse samples of artificial graphite are represented graphically in Figures 1 and 2, respectively. The curves in these figures are plotted from different origins to display the individual characteristics of each curve. If drawn from the same origin the curves would be so close together as to be confusing. They do not show any irregularities. After cooling to room temperature the change in length from the original length before heating did not exceed 0.012 per cent in any test.

The average coefficients of expansion given in Table 3 were derived from the expansion curves of Figures 1 and 2.

TABLE 3.—*Average coefficients of expansion of artificial graphite*

Sample	Carbon content	Average coefficients of expansion per degree centigrade						Test No.
		20 to 100° C.	20 to 200° C.	20 to 300° C.	20 to 400° C.	20 to 500° C.	20 to 600° C.	
Longitudinal:	*Per cent*	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	
1180A	99.45	1.9	2.0	2.2	2.4			1
		1.9	2.0	2.2	2.4	2.6		2
				2.1		2.5	2.7	3
1181	99.12	2.2	2.1	2.3	2.4			1
1182	99.72	1.8	1.9	2.1	2.3			1
Transverse:								
1183	99.16	3.1	3.2	3.4	3.6			1
1184	99.35	2.8	3.0	3.2	3.4			1
		2.9	2.8	3.1	3.4	3.6	3.7	2

An examination of Table 3 shows the following interesting facts: The coefficients of expansion of graphite are low.[2] For example, the coefficient of expansion of ordinary steel is about six times the coefficient of longitudinal sections of graphite and about four times the value for transverse sections between 20 and 100° C. It is evident that the transverse samples expand considerably more than the longitudinal samples. The coefficients of expansion of all samples increase with temperature. For both the longitudinal and transverse samples the coefficients of expansion decrease slightly

[2] Graphite has been found to exhibit resistance to sudden temperature changes. Plunging samples at 600° C. into cold water (10° C.) did not cause breakage or cracking.

as the purity (carbon content) increases. The coefficients of samples of small cross section (see Table 2, samples 1180A and 1184) do not differ materially from those of samples of larger cross section. The coefficients of expansion on a second or third heating agree well with the values obtained on the first heating.

Figure 3 shows the average expansion curves of longitudinal and transverse graphite, and Table 4 shows a comparison of the average coefficients of expansion. These average curves and average coefficients of expansion were obtained from the data on all samples investigated.

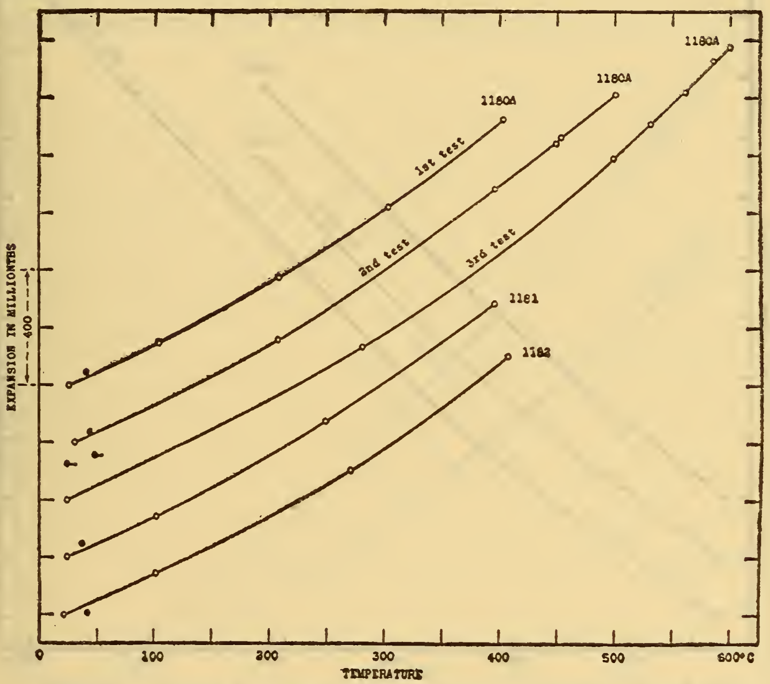

Fig. 1.—*Linear expansion of longitudinal samples of artificial graphite*

The light circles represent observations on heating and the dark circles represent observations on cooling. For the third test of sample 1180A, the tagged circles represent observations on cooling.

Table 4.—*Comparison of average coefficients of expansion of longitudinal and transverse samples of artificial graphite*

Material	Average coefficients of expansion per degree centigrade					
	20 to 100° C.	20 to 200° C.	20 to 300° C.	20 to 400° C.	20 to 500° C.	20 to 600° C.
	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$
Longitudinal	1.9	2.0	2.2	2.4	2.5	2.7
Transverse	2.9	3.0	3.2	3.5	3.6	3.7

The coefficients of expansion of transverse graphite exceed those of longitudinal graphite by 45 per cent, approximately. Other investigators have obtained similar results in the case of wood, but to a more marked extent. Hendershot [3] found that the expansion of some woods across the fiber is 4 to 17 times the expansion of these woods parallel to the fiber. The tensile strength [4] of longitudinal graphite has been reported [5] to be about 65 per cent greater than the value for the transverse graphite.

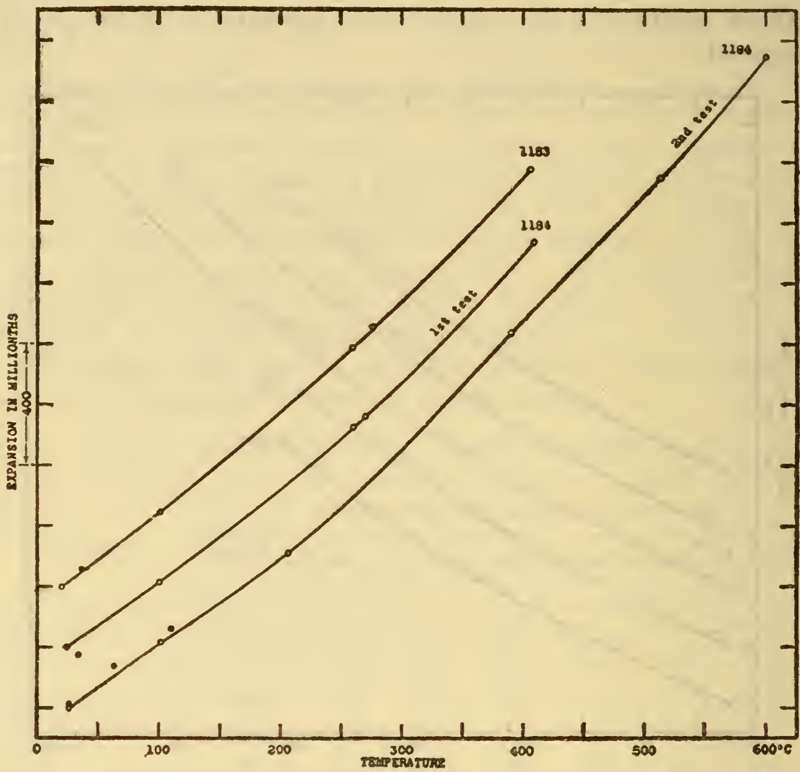

FIG. 2.—*Linear expansion of transverse samples of artificial graphite*

The light circles represent observations on heating and the dark circles represent observations on cooling.

The differences [6] in expansion and tensile strength between the transverse and longitudinal graphite may be due to the fact that the

[3] Hendershot, Science, **60**, p. 456; Nov. 14, 1924.

[4] 800 to 1,000 lbs./in.^{2} for longitudinal, and 500 to 600 lbs./in.^{2} for transverse graphite.

[5] Catalogue of Acheson Graphite Co.

[6] C. E. Taylor, of the Acheson Graphite Co., states that further consideration of the effects of the extrusion process by which the "green" electrodes are formed will be helpful. During extrusion the outer portions are retarded by friction with the walls of the die. There is therefore a good deal of internal motion parallel to the axis of the forming electrode. This internal motion causes a marked difference in the structure in the two directions. There is a tendency to a laminated structure in the transverse direction. The aggregate particles and cell walls are more firm and continuous in a lengthwise direction of the electrode than in a crosswise direction. Voids or pores tend to form with their longer axes lengthwise of the electrode. It seems quite probable that this has an effect on expansivity in the two directions. The longitudinal increment is practically a measure of the linear expansion of the material, and the transverse change is the linear expansion plus an increment of distortion due to increased spring in the arches formed by the cell walls of the voids and pores. Taylor also believes that the lower tensile strength in the transverse direction is due to the laminated structure mentioned.

graphite in the process of manufacture has been compressed differently in the two directions.

From the results of previous investigations (see Table 1) it is seen that the coefficients of expansion of various forms of carbon vary considerably. The only data previously available on artificial graphite were those published by Day and Sosman [7] in 1912 and by Sieglerschmidt [7] in 1920. The coefficients of expansion reported by the former are considerably less than the values obtained in the present investigation. Sieglerschmidt's value for the range from 0° to 250° C. is in agreement with the results given by the present authors for transverse graphite.

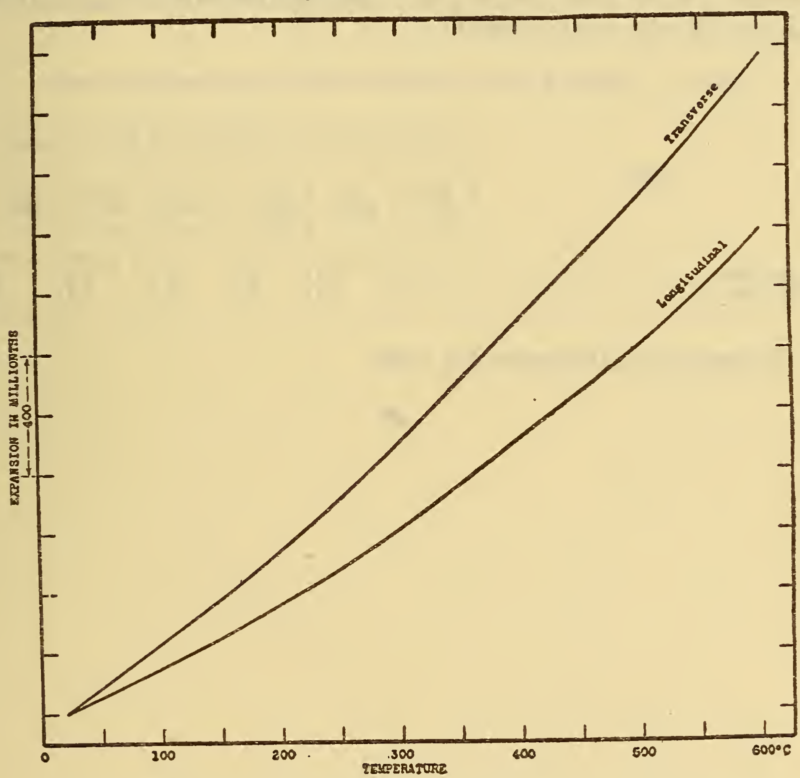

Fig. 3.—*Average expansion curves of artificial graphite (longitudinal and transverse directions)*

V. SUMMARY

This paper gives the results of an investigation on the thermal expansion of longitudinal and transverse samples of artificial graphite (99.2 to 99.7 per cent carbon) over various temperature ranges between room temperature and 600° C.

A summary of available data by previous observers on the thermal expansion of graphite (natural and artificial) and of other forms of carbon is included.

[7] See Table 1.

The coefficients of expansion of graphite are low. For example, the coefficient of expansion of ordinary steel is about six times the coefficient of longitudinal sections of graphite and about four times the value for transverse sections between 20 and 100° C. The transverse samples expand considerably more than the longitudinal samples (approximately 45 per cent). The coefficients of expansion of all samples increase with temperature. For the longitudinal and transverse samples the coefficients of expansion decrease slightly as the purity (carbon content) increases.

The following table gives a résumé of average coefficients of expansion derived from the data on all samples for various temperature ranges between 20 and 600° C.:

TABLE 5.—*Résumé of average coefficients of expansion of artificial graphite*

Material	Average coefficients of expansion per degree centigrade					
	20 to 100° C.	20 to 200° C.	20 to 300° C.	20 to 400 °C.	20 to 500° C.	20 to 600° C.
	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$
Longitudinal	1.9	2.0	2.2	2.4	2.5	2.7
Transverse	2.9	3.0	3.2	3.5	3.6	3.7

WASHINGTON, December 16, 1926.